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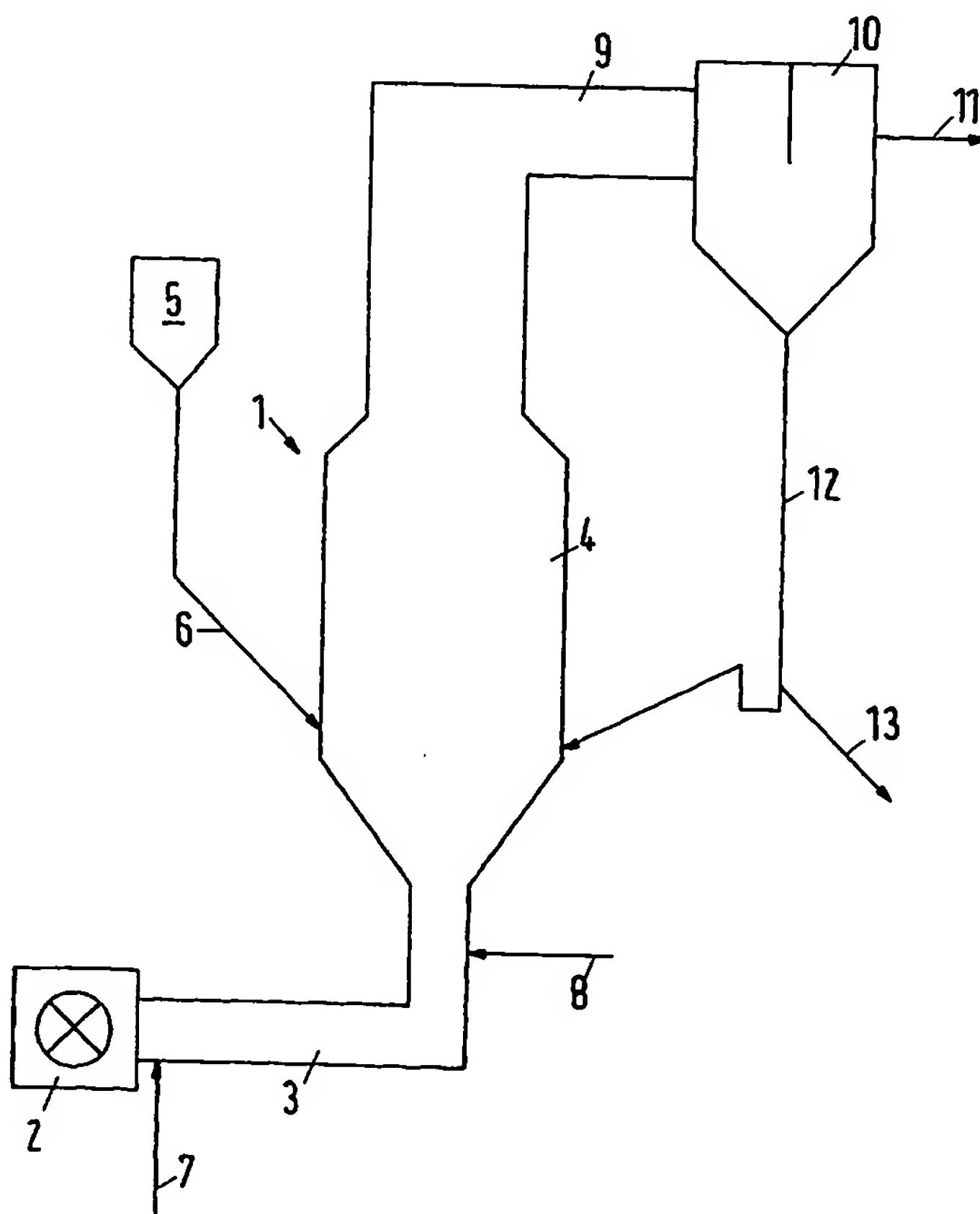
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(54) Title: METHOD AND PLANT FOR THE THERMAL TREATMENT OF GRANULAR SOLIDS



(57) Abstract: This invention relates to a method for the thermal treatment of granular solids in a reactor (1) with swirl chamber (4), which in particular constitutes an flash reactor or suspension reactor, wherein the microwave radiation from a microwave source (2) is fed into the reactor (1) through a wave guide, and to a corresponding plant. To avoid deposits in the wave guide, the same constitutes a gas supply tube (3), a gas stream being additionally fed through the gas supply tube (3) into the swirl chamber (4).



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## METHOD AND PLANT FOR THE THERMAL TREATMENT OF GRANULAR SOLIDS

### Technical Field

This invention relates to a method for the thermal treatment of granular solids in a reactor with a swirl chamber, which in particular constitutes an flash reactor or suspension reactor, wherein microwave radiation is fed into the reactor through at least one wave guide, and to a corresponding plant. In this method, granular solids are thermally treated in a fluidized bed formed in the reactor, wherein fluidizing gas and electromagnetic waves (microwaves) coming from a microwave source are fed into the fluidized bed of the reactor, which constitutes a fluidized layer.

There are several possibilities for coupling a microwave source to such fluidized-bed reactors. These include for instance an open wave guide, a slot antenna, a coupling loop, a diaphragm, a coaxial antenna filled with gas or another dielectric, or a wave guide occluded with a microwave-transparent substance (window). The type of decoupling the microwaves from the feed conduit can be effected in different ways.

Theoretically, microwave energy can be transported in wave guides free of loss. The wave guide cross-section is obtained as a logical development of an electric oscillating circuit comprising coil and capacitor towards very high frequencies. Theoretically, such oscillating circuit can likewise be operated free of loss. In the case of a substantial increase of the resonance frequency, the coil of an electric

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oscillating circuit becomes half a winding, which corresponds to the one side of the wave guide cross-section. The capacitor becomes a plate capacitor, which likewise corresponds to two sides of the wave guide cross-section. In reality, an oscillating circuit loses energy due to the ohmic resistance in coil and capacitor.

- 5 The wave guide loses energy due to the ohmic resistance in the wave guide wall.

Energy can be branched off from an electric oscillating circuit by coupling a second oscillating circuit thereto, which withdraws energy from the first one. Similarly, by flanging a second wave guide to a first wave guide energy can be decoupled from the same (wave guide transition). When the first wave guide is shut off behind the coupling point by a shorting plunger, the entire energy can even be diverted to the second wave guide.

- 10 15 The microwave energy in a wave guide is enclosed by the electrically conductive walls. In the walls, wall currents are flowing, and in the wave guide cross-section an electromagnetic field exists, whose field strength can be several 10 KV per meter. When an electrically conductive antenna rod is put into the wave guide, the same can directly dissipate the potential difference of the electromagnetic field and with a suitable shape also emit the same again at its end (antenna or probe decoupling). An antenna rod which enters the wave guide through an opening and contacts the wave guide wall at another point can still directly receive wall currents and likewise emit the same at its end. When the wave guide is shut off behind the antenna coupling by a shorting plunger, the entire energy 20 25 can be diverted from the wave guide into the antenna in this case as well.

When the field lines of the wall currents in wave guides are interrupted by slots, microwave energy emerges from the wave guide through these slots (slot decoupling), as the energy cannot flow on in the wall. The wall currents in a rectangular wave guide flow parallel to the center line on the middle of the broad

side of the wave guide, and transverse to the center line on the middle of the narrow side of the wave guide. Transverse slots in the broad side and longitudinal slots in the narrow side therefore decouple microwave radiation from wave guides.

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Microwave radiation can be conducted in electrically conductive hollow sections of all kinds of geometries, as long as their dimensions do not fall below certain minimum values. The exact calculation of the resonance conditions involves rather complex mathematics, as the Maxwell equations (unsteady, nonlinear differential equations) must ultimately be solved with the corresponding marginal conditions. In the case of a rectangular or round wave guide cross-section, however, the equations can be simplified to such an extent that they can be solved analytically and problems as regards the design of wave guides become clearer and are easier to solve. Therefore, and due to the relatively easy production, only rectangular wave guides or round wave guides are used industrially, which are also preferably used in accordance with the invention. The chiefly used rectangular wave guides are standardized in the Anglo-Saxon literature. These standard dimensions were adopted in Germany, which is why odd dimensions appear in part. In general, all industrial microwave sources of the frequency 2.45 GHz are equipped with a rectangular wave guide of the type R26, which has a cross-section of 43 x 86 mm. In wave guides, different oscillation states exist: In the transversal electric mode (TE mode), the electric field component lies transverse to the wave guide direction and the magnetic component lies in wave guide direction. In the transversal magnetic mode (TM mode), the magnetic field component lies transverse to the wave guide direction and the electric component lies in wave guide direction. Both oscillation states can appear in all directions in space with different mode numbers (e.g. TE-1-1, TM-2-0).

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A method for the thermal treatment of granular solids is known from US 5,972,302, wherein sulfidic ore is subjected to an oxidation supported by microwaves. This method is chiefly concerned with the calcination of pyrite in a fluidized bed, wherein the microwaves introduced into the fluidized bed promote the formation of hematite and elementary sulfur and suppress the formation of SO<sub>2</sub>. There is employed a stationary fluidized bed which is directly irradiated by the microwave source disposed directly above the same. The microwave source or the entrance point of the microwaves necessarily gets in contact with the gases, vapors and dusts ascending from the fluidized bed.

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EP 0 403 820 B1 describes a method for drying substances in a fluidized bed, wherein the microwave source is disposed outside the fluidized bed and the microwaves are introduced into the fluidized bed by means of a wave guide. There are frequently reflections of microwave radiation at the solids to be heated, whereby the efficiency is reduced and the microwave source is possibly damaged. In the case of open microwave wave guides, there are also dust deposits in the wave guide, which absorb part of the microwave radiation and can damage the microwave source.

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### Summary of the Invention

It is therefore the object underlying the invention to make the feeding of microwaves into a stationary or circulating fluidized bed more efficient and protect the microwave source against the resulting gases, vapors and dusts and the reflected microwave power.

In accordance with the invention, this object is substantially solved in a method as mentioned above in that the wave guide constitutes a gas supply tube and that in addition to the microwave radiation a gas stream is fed into the swirl chamber through the gas supply tube.

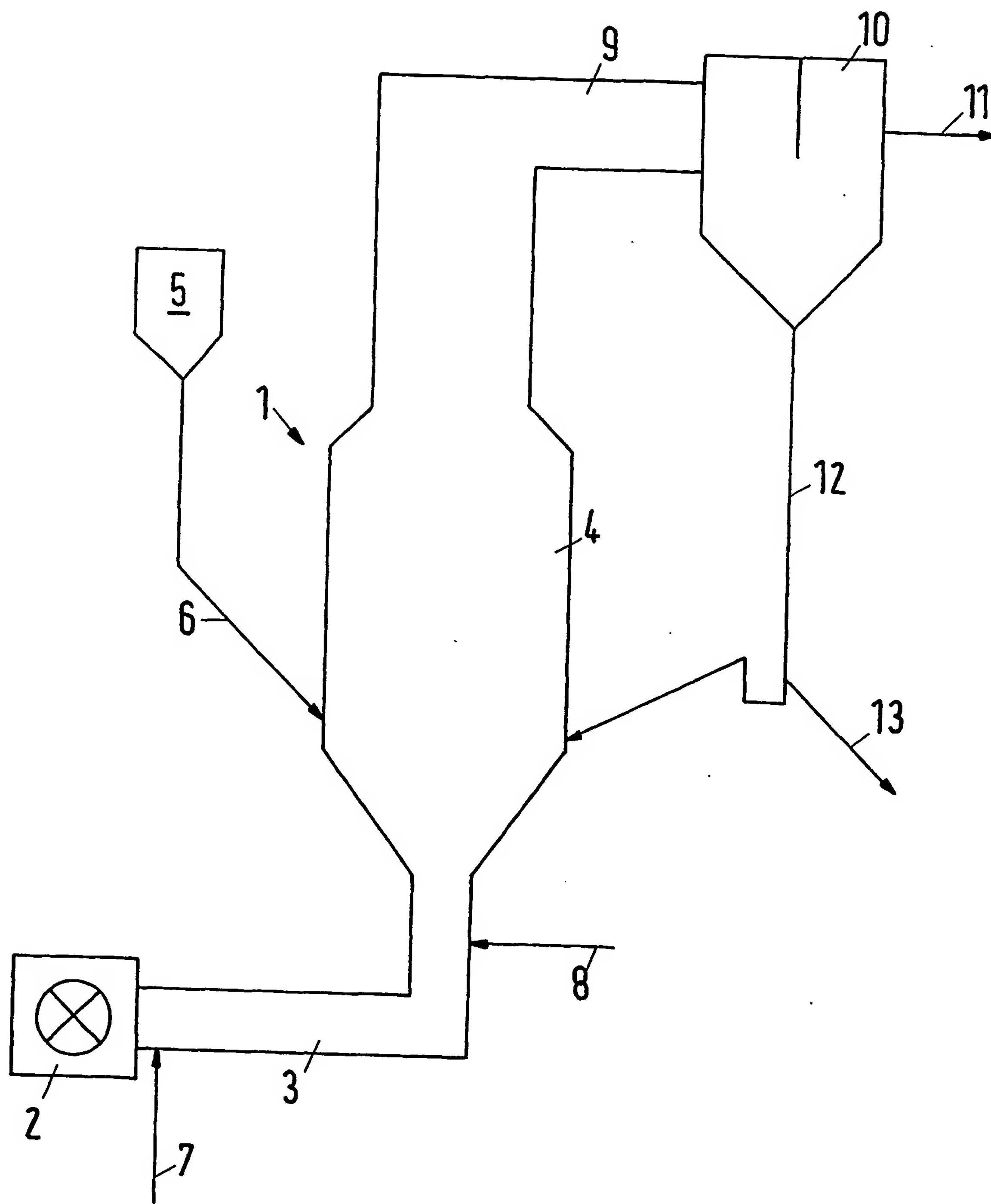
- 5 -

By means of the continuous gas stream from the wave guide it is reliably avoided that dust or process gases enter the wave guide, spread up to the microwave source and damage the same or form solid deposits in the wave guide.

5 In accordance with the invention, microwave-transparent windows in the wave guide for shielding the microwave source, as they are commonly used in the prior art, can therefore be omitted. The same involve the problem that deposits of dust or other solids on the window can impair and partly absorb the microwave radiation. Therefore, the open wave guides in accordance with the invention are particularly advantageous. Thus, the microwave source can be arranged outside the circulating fluidized bed, the microwave radiation being fed into the fluidized-bed reactor through at least one open wave guide together with a gas stream.

15 It is also possible to introduce still dust-laden, hot process gas through the gas supply tube constituting a central tube or central gas tuyere into the reactor, with which process gas the solids in the swirl chamber are swirled. Since dust-laden gas would, however, reduce the efficiency of the microwave irradiation due to the absorption of microwave radiation by the dust particles, neutral, dust-free  
20 gas, e.g. purge gas, would first be passed through the gas supply tube in accordance with the invention, which neutral gas does not react with the substances contained in the reactor and hardly absorbs the microwave radiation. In continuation of this inventive idea, the dust-laden process gas is only introduced into the reactor space shortly before the entrance of the gas supply tube (central  
gas tuyere). During the thermal treatment in the circulating fluidized bed of the reactor, the solids circulate continuously between a fluidized-bed reactor (flash  
25 or suspension reactor), a solids separator connected with the upper region of the reactor, and a return conduit connecting the solids separator with the lower region of the fluidized-bed reactor. Usually, the amount of solids circulating per

Fig.1



## INTERNATIONAL SEARCH REPORT

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## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)  
EPO-Internal, WPI Data, PAJ

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category •	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3 528 179 A (SMITH FRANKLIN J) 15 September 1970 (1970-09-15) column 4, line 43 -column 5, line 20 figure 6	1-4,6,7
A	WO 98 08989 A (EMR MICROWAVE TECHNOLOGY CORP) 5 March 1998 (1998-03-05) page 6, paragraph 1 -page 7, paragraph 2 figure 1	1,4,6

 Further documents are listed in the continuation of box C. Patent family members are listed in annex.

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## INTERNATIONAL SEARCH REPORT

Information

A patent family members

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Document No

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Patent document cited in search report		Publication date	Patent family member(s)		Publication date
US 3528179	A	15-09-1970	NONE		
WO 9808989	A	05-03-1998	AU	721221 B2	29-06-2000
			AU	3843997 A	19-03-1998
			CA	2263497 A1	05-03-1998
			WO	9808989 A1	05-03-1998
			EP	0950122 A1	20-10-1999
			US	5972302 A	26-10-1999
			ZA	9707567 A	22-02-1999